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ENVIRONMENTAL HEALTH SERIES
Radiological Health

SOIL AND SEDIMENT ANALYSIS: PREPARATION OF SAMPLES FOR ENVIRONMENTAL RADIATION SURVEILLANCE

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ENVIRONMENTAL HEALTH SERIES

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SOIL AND SEDIMENT ANALYSIS: PREPARATION OF SAMPLES FOR ENVIRONMENTAL RADIATION SURVEILLANCE

Part I—Preparation of Soil Samples:
Laboratory Techniques

John H. Abrahams, Jr.

Part II—Ultrasonic Dispersion of
Sediments for Size Fraction Radioanalysis

Raymond H. Johnson, Jr.
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ABSTRACT

Soil samples were separated into size fractions to determine those in which uptake was greatest following the addition of different concentrations of zinc 65 tracer in aqueous suspensions, and to evaluate several methods of fractionation. Activity per gram for particles less than 43 microns was 7 to 8 times greater than for 43 to 250 micron particles. This factor increased as the particle sizes decreased. Activity on each size fraction increased with increasing concentrations of the zinc 65 tracer in aqueous suspensions, as well as with increasing organic content. Wet sieving with mild physical dispersion was a more convenient and reproducible method of separating the fine and coarse fractions than sedimentation and elutriation.

Ultrasonic vibrations and high-speed stirring were evaluated as means for achieving the dispersion necessary prior to screening for radioanalysis of sediment fractions. A microscopic analysis of sediments before and after dispersion indicated that ultrasonic treatment released silt particles bound together by organic matter. The degree of dispersion was found to be generally dependent on sonification time but samples of about 35 grams were adequately dispersed after 15 to 20 minutes of treatment. In contrast, high-speed stirring caused little dispersion during the same time interval.

Ultrasonic dispersion and screening of sediments dosed with the gamma emitter zinc 65 indicated that 54 percent of the activity was associated with particles smaller than 43 microns. A corresponding fraction after stirring contained only 32 percent of the activity. This demonstrates the large errors possible owing to poor dispersion by stirring.

These studies demonstrate that bulk samples composed largely of coarse material generally are poor indicators of radionuclide concentrations. By counting that fraction of the field samples, which contains the predominant amount of activity, and by standardizing laboratory procedures, the usefulness and sensitivity of these samples for surveillance would be greatly enhanced.

INTRODUCTION

Early and accurate detection of environmental radiocontamination becomes increasingly important with the growing trend toward construction of nuclear facilities near population centers. Thus, reliance on time and distance to reduce potential hazards of the radionuclides discharged is decreased substantially, and reliable methods are needed to detect low levels of radiocontamination as well as accidental releases.

Ordinarily, only low-level liquid wastes are released to the terraqueous environment. When liquid wastes are discharged to seepage pits above the ground water table, into surface waters near the sites, or into ravines or sewers that lead to either of these areas, radionuclides may be sorbed by earth materials and biota. Such components are either deposited as bottom sediment or dispersed into watercourses.

Soil, sediment, and water samples are collected at sites downstream from waste disposal sites for evaluation of the radionuclide content. Water samples, particularly those from less turbulent streams, tend to reflect the contamination level at the moment of collection, but soil and sediment samples may indicate some recent past as well as present radiocontamination levels. Therefore, soil and sediment samples may show evidence of contamination not detected in water samples.

For surveillance studies, comparison of activity levels in soil and sediment samples is usually difficult, particularly for "grab" samples, because of the wide variability in composition of both soil and sediment samples. Thus, radioactivity levels of sediment samples, for example, are likely to be a function of sample composition rather than of the amount of radioactivity present in the ambient water.

For this reason, care at each step of the procedure—sample collection, preparation, and analysis—is vital for quantifying the radionuclide content of geologic and hydrologic materials. These studies were conducted to determine whether some active portion of a "grab" sample, easily separated from the inert part of the sample, would provide a sensitive and reproducible indicator of radiocontamination in the environment. Although the experimental material for these studies consisted of soil and sediments from eastern Massachusetts, it is believed that, when standardized, the method will provide a useful and sensitive indicator for monitoring bottom sediments in general.

PART I

PREPARATION OF SOIL SAMPLES: LABORATORY TECHNIQUES

John H. Abrahams, Jr.

In order to follow the migration of radionuclides *(1, 2)*, samples of soil and fill material are collected beneath and adjacent to seepage pits receiving radioactive wastes. These samples are often processed and evaluated with little regard for soil properties, which generally vary widely with location, soil type, and depth of sampling *(3, 4)*. These properties are important because observed differences in concentrations of radioactivity may reflect variations in the physical and chemical composition of the sample.

Factors that may affect uptake of radionuclides by the soil include particle size, the kind of clay minerals present *(5, 6, 7, 8)*, the chemical nature of the soil, and the kind and amount of organic matter present *(9, 10, 11)*. Evaluation of some of these variables generally requires complex laboratory determinations; therefore, an alternative was sought in laboratory studies of soil aimed at developing a simplified standard method based on uptake characteristics of zinc 65 related to the size of particles comprising the samples. For this study, samples of a composite New England soil were separated into size fractions so that the activity of a particular particle size could be determined accurately. During separation of these fractions, emphasis was placed on physical fractionation in order that the chemical equilibria of the soil suspension would not be disturbed nor the sorption affected.

Even though uptake is known to be inversely proportional to particle size *(7)*, it is not known whether these smaller sizes are the best means of making direct comparison between samples. Thus, other objectives of this study were to determine the size fraction of a soil with an uptake capacity most responsive to different concentrations of zinc 65 in aqueous suspensions, and to evaluate methods of separation for obtaining this fraction for radiocontamination studies. Because arbitrary procedures may result in values that are difficult to relate to field conditions, another purpose was to standardize procedures from sample collection through laboratory preparation and gamma counting.

MATERIALS AND METHODS

The soil collected for this study was a composite of the upper 3 feet of a Gloucester stony fine sandy loam, which was derived from glacial till composed mostly of granite and gneiss *(12)*. To separate the weaker soil aggregates, the soil was air dried, then sieved mechanically for 30 minutes on screens with openings ranging from 250 to 43 microns. Oven drying at elevated temperatures would have increased the stability of the aggregates and made physical separation into size fractions more difficult. These soil or particle aggregates form the main structure of most soils. They generally consist of various sizes of organic and primary particles bound by cementing agents such as organic colloids or iron oxides. Some of these aggregates, however, disperse in water, whereas others are water stable but disperse during mild physical or chemical treatment.

After dry sieving, equal portions of each size fraction were recombined into a composite soil. The organic matter of this composited soil was 6.7 percent as measured by modifications of the Walkley-Black chromic acid oxidation method *(13, 14)*. The cation exchange capacity was 10.8 milliequivalents per 100 grams as measured by the barium displacement method *(1, 15)*. The sand content ($>50\mu$) was about 50 percent, and the clay ($<2\mu$), about 2 percent.

In preparing each sample, the composited soil was initially stirred for 10 minutes in deionized water maintained at a pH range of 6.3 to 7.3. Stirring longer than this produced no further reduction in the percentage of larger aggregates.

FRACTIONATION PROCEDURES

After initial preparation, each sample was fractionated by the common methods of wet sieving, elutriation, or sedimentation *(5, 2)*, and by combinations of these methods. Preliminary studies were conducted without using radioactive materials. Next, studies using zinc 65 as a tracer ion followed for most of the procedures developed.

During wet sieving, the soil was separated on a 43-micron screen into a size fraction less than 43 microns and a coarse fraction of 43 to 250 microns by washing with a small stream of water. Further separation was obtained by rubbing the soil on the screen with a rubber policeman until the water passing through the screen was clear.

Sedimentation and elutriation procedures were used for particles down to 20 microns and smaller *(5, 16, 17, 18)*. In the sedimentation technique, the sample was allowed to settle through a 2-inch column of water in a glass cylinder about 40 inches long. Turbulence created by introducing the sample to the water was reduced by using a rubber disk with brass fittings that was positioned near the top of the water. Lifting one side of the disk slowly allowed

the sample placed on top to slip off the low side. After a prescribed settling time determined by Stokes law *(5)*, aliquots of the soil suspension containing different size fractions were withdrawn through the tapered bottom of the cylinder. Each aliquot was examined for weight, organic matter, and particle size. Several different sample sizes and settling times were used. Particle sizes were determined directly by microscopy.

In the elutriation procedure, a current of water was directed upward through a series of vertical columns with successively larger diameters. Both water and sample were introduced through the bottom of the smallest column. The smaller particles tended to move upward with the water, but the larger particles remained in the smaller columns containing the higher velocities. Particles in the size range desired were collected in the larger columns.

For the elutriation study, radioactive material was not used because the fractionation by elutriation was cumbersome and required critical control of the flow through the columns. Furthermore, the equipment seemed inadequate for separating smaller particles and the large quantity of water needed to separate the smaller size fractions was difficult to handle. Even with these disadvantages, however, size separations at about 50, 40, 30, 20, and 10 microns were possible, but the process was abandoned for more convenient methods.

TRACER STUDY PROCEDURES

Zinc 65 was chosen as a tracer ion because it occurs commonly in liquid nuclear wastes *(2)*, and because it is easily measured by gamma spectrometry. Furthermore, it is readily sorbed on soils and on other earth materials in the middle pH range *(8)*, and has a relatively long half-life of 245 days. In this study, the zinc 65 concentration was below 10^{-4} microcuries per liter, the maximum permissible concentration (MPC) in water *(19)*.

The soil fractions obtained were counted in plastic containers 4 inches in diameter at a standard volume of 40 milliliters, with a 4- by 4-inch NaI (Tl) crystal connected to a multichannel gamma spectrometer. Calibration curves were prepared relating counting efficiency to sample size.

Two series of tracer studies were conducted with 20-gram samples of composited soil in small glass containers. In the first study, the soil samples were suspended in 200 milliliters of water in 400-milliliter beakers, and the suspensions dosed with five different concentrations of zinc 65. After stirring for 5 minutes, the suspensions were allowed to settle overnight before being separated by wet sieving. In the second study, 20 liters of water containing zinc 65 were divided evenly among four tanks, three containing soil and one serving as a

control. After stirring and settling overnight, each suspension was separated by filtration through a 43-micron screen and filter paper. Duplicate samples of coarse and fine fractions were analyzed by gamma spectrometry.

Samples used in the sedimentation studies were first dosed with zinc 65 in small glass containers, as in the wet-sieving studies. The dosed samples were then separated into size fractions by the sedimentation process, and each fraction was counted for radioactivity.

RESULTS

The wet-sieving studies made without zinc 65 activity showed that increasing the degree of agitation caused a higher percentage of the sample to pass a 43-micron screen. As shown in table 1, about 30 percent of the sample washed through the screen without treatment. Further agitation, by either rubbing and washing, or stirring and washing, caused additional breakage of particle aggregates, so that an increased amount of particles passed through the screen. Only minor additional dispersion and separation were observed after stirring in a dispersing agent.

Table 1. Effect of mild physical treatment on fractionation by elutriation and sieving

Treatment	Percentage > 43 microns[a]
Elutriation only	74
Using 43-micron screen	
Wash only	70
Stir and wash	67
Stir and rub	55
Dispersing agents,[b] and rub	49

[a] ±0.5%
[b] NH_4OH or NaOH

Results of radioactive tracer experiments plotted in figure 1 indicate that the zinc 65 sorbed per gram of the fine fraction was 7 to 8 times that sorbed by the coarse fraction. Further agitation such

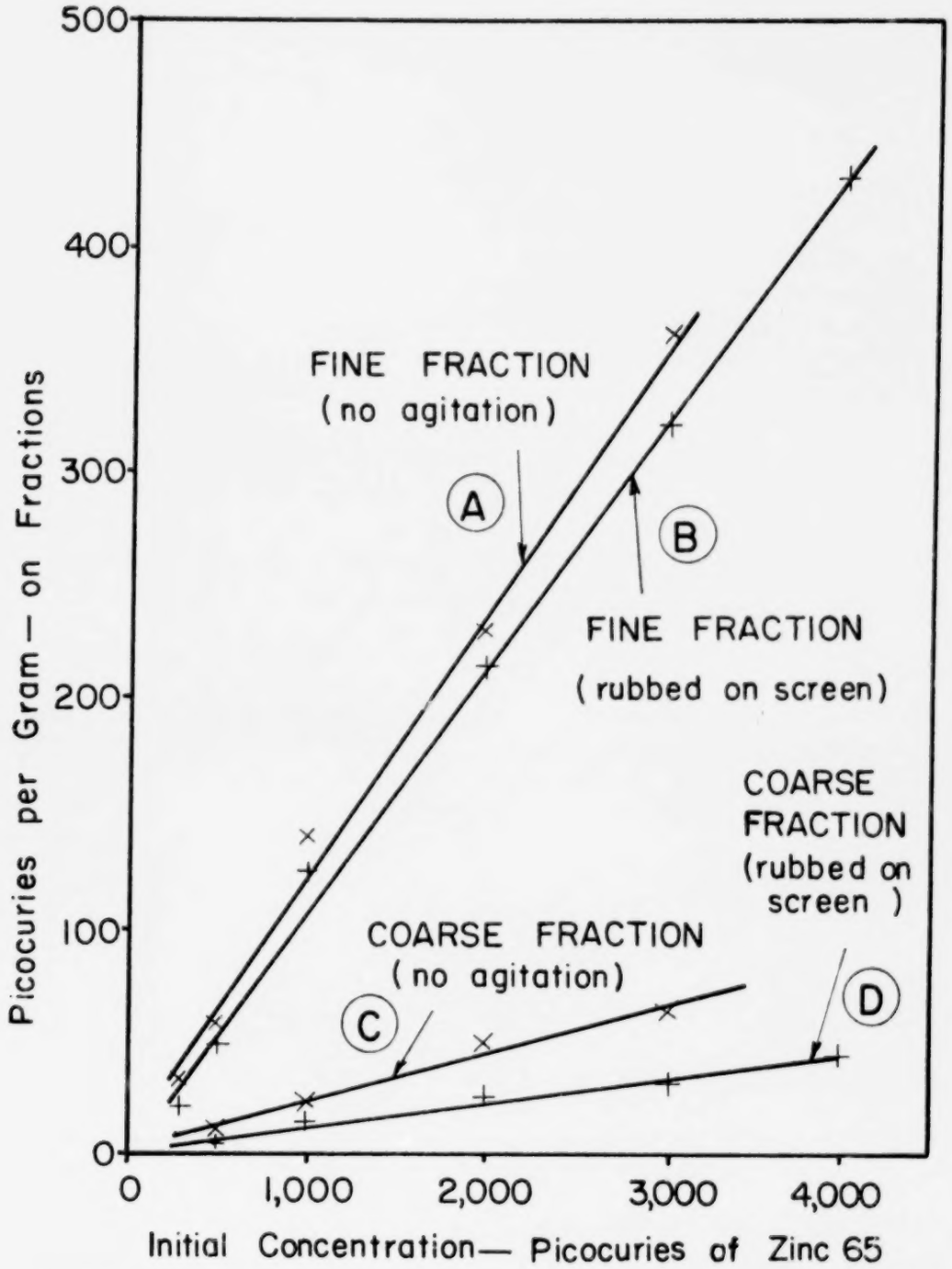

Figure 1. Zinc 65 in fine and coarse soil fractions separated from suspensions in solutions of different zinc 65 concentrations. (Fine fraction includes all sizes below 43 microns.)

as rubbing the soil during wet sieving, however, removed additional material containing the zinc 65 from the coarse fraction, as illustrated by curves C and D. This material separated by rubbing then became part of the fine fraction, but the zinc 65 activity on a weight basis was less on this fraction as shown by curves A and B. Therefore, this portion probably consisted of relatively inert silt size particles separated by breaking the cement bond holding the larger aggregates together.

This study also showed that zinc 65 activity sorbed on either fraction was directly related to the initial concentration of zinc 65 in suspensions. This demonstrates the reproductibility of the wet-sieving method. In addition, the relatively greater amounts of zinc 65 activity sorbed on the fine fraction suggests that this fraction would be the most useful in surveillance studies.

The second study, summarized in table 2, showed that the soil removed more than 98 percent of the zinc 65 from the water. Only a negligible amount of activity was sorbed on the plastic lining of the tanks containing the soil, compared to about 10 percent on the lining in the control tank. The ratio of zinc 65 retained by the fine fraction to zinc 65 retained by the coarse fraction was about 7 to 1.

Table 2. Distribution of zinc 65 in soil suspensions [a]

Sample	Gamma activities of separated fractions and filtrate	
	Control tank	Average for 3 soil suspension tanks
Filtrate (5 liters), pCi/liter	4,950	180
Coarse fraction (>43 microns), pCi/g	–	375
Fine fraction (<43 microns), pCi/g	–	2,655
On plastic liner of tanks, pCi	2,750	<20

[a] Dosage 5,500 pCi zinc 65 per liter.

DISTRIBUTION OF ACTIVITY BY PARTICLE SIZE

Zinc 65 activity on a weight basis associated with the size fractions obtained by sedimentation increased almost linearly with decreasing particle diameter, and was almost directly proportional to the initial activity. This trend, shown on figure 2, was evident for all four samples used, even though fractionation procedures were slightly different. The slight deviation of the values from the straight lines plotted on figure 2 is attributed largely to the fraction

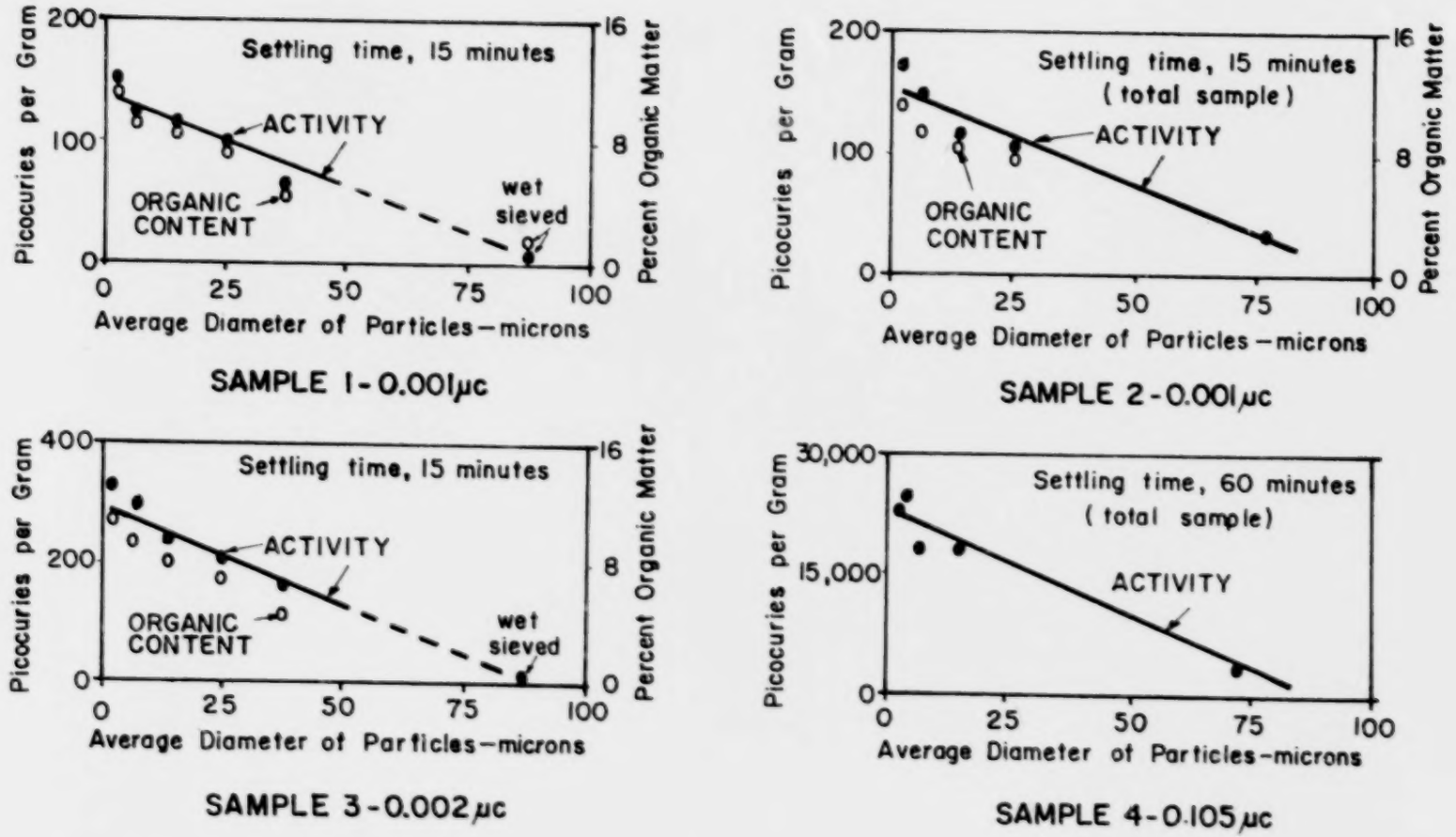

Figure 2. Zinc 65 activity and organic matter content of various size fractions of composited soil after sedimentation

less than 5 microns, which probably was mostly clay and organic particles. Furthermore, there seems to be a general correlation between the sorbed activity and the percentage of organic matter for each of the size fractions (fig. 2). This suggests that the amount of organic matter as well as the surface area of the silt and clay particles apparently was a factor controlling uptake on the sandy soil.

The activity on each of the smaller size fractions was low because of the small sample sizes produced by this subdivision by sedimentation. The weight of these fine fractions would be even smaller for samples composed mostly of coarse material. Despite these disadvantages, however, the process of obtaining fractions smaller than 43 microns was relatively simple as well as accurate and reproducible, and would be useful for research on surveillance of soil samples containing radioactivity.

A brief study with an organic sediment sample collected from Fairhaven Bay on the Sudbury-Concord River demonstrated some difficulties encountered when dispersing a sample chemically rather than physically. Chemical dispersion was used because this material, about 31 percent organic matter, could not be fractionated satisfactorily by wet sieving. A sample dosed with zinc 65 was boiled in hydrogen peroxide (30%) for 1 hour, and allowed to settle overnight. About 88 percent of the activity was in the supernatant liquid. However, because of the pretreatment required for this sample, it was difficult to determine whether this activity was originally contained on the organic matter, or if some had been removed from the solids.

CONCLUSIONS AND RECOMMENDATIONS

Laboratory studies of fractions of a New England soil indicated that uptake of zinc 65 tracer by particles smaller than 43 microns was 7 to 8 times the uptake by particles ranging from 43 to 250 microns. Uptake of zinc 65 on a "grab" sample was confirmed to occur mostly on a relatively small portion of the total sample. Thus, this portion could be separated from the bulk of the sample for use in surveillance studies.

Wet sieving coupled with mild physical treatment of soil samples was found to be the most convenient of the methods tested for size fractionation, as well as the most reproducible. The processes of sedimentation and elutriation, in which particles as small as 5 microns were separated, also could be standardized, but were less convenient than the wet-sieving method.

The direct relation observed between the zinc 65 activity sorbed on the fine fraction and the initial zinc 65 concentration indicates that with proper standardization of the wet-sieving method, along with standard analytical techniques, it should be possible to compare quantitatively the radioactivity in one soil sample with that in another. With corrections for the composition of samples, differences between activity levels would be indicative of the activity of the environment from which they were taken. Although zinc 65 and the soil fraction less than 43 microns were used in this study, these procedures may be applicable to other gamma emitters and size ranges, and should be investigated further.

These studies also show that because of the apparent correlation between activity and organic matter, the organic matter associated with the smaller size fraction could be used to compare the concentration of radionuclides sorbed on soil materials. Because of difficulties encountered in separating the organic fraction from the solids by physical means, however, some other method of correlating the organic fraction and activity may be needed. This is particularly important where the organic fraction and the clay content are high.

In conclusion, these investigations showed that in soils and sediments, since the radioactivity probably was associated with only a small fraction of the sample solids, particularly the organic, clay, and silt portions, a substantial improvement in radioassay might be achieved by first separating these from the coarse solids. This fine material appeared to be the main fraction of sediment solids representative of the suspended matter in streams on which the radionuclide was sorbed. Physical rather than chemical separation of fractions is suggested because physical separation would not change the equilibria in the sample.

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PART II

ULTRASONIC DISPERSION OF SEDIMENTS FOR SIZE FRACTION RADIOANALYSIS

Raymond H. Johnson, Jr., and John H. Abrahams, Jr.

Sampling and analysis of bottom sediments are becoming increasingly important in determining the distribution of radioactive wastes in an aquatic environment. The usefulness of sediment analyses, however, depends on the standardization of laboratory procedures for measuring radioactivity that will allow comparison of samples of widely varying compositions. Since particle size is a primary factor in sediment composition, a study was initiated to develop a procedure for accurate size fractionation of sediments. In particular, ultrasound was evaluated as a means of physically dispersing sediment particles prior to screening for radioanalysis of size fractions.

LITERATURE REVIEW

The analysis of sediments for radioactivity necessitates an understanding of the manner in which activity is associated with sediments. Sayre, Guy, and Chamberlain *(1)*, have noted that most radioactive wastes discharged into natural waterways tend to become sorbed on suspended solids which settle to form bottom sediments. The amount of sorption depends on the type of sediment, the radionuclide being sorbed, the available surface area, and the amount and type of dissolved materials in the water. They have also noted that, of these factors, surface area as determined by particle size was the most important single sediment characteristic governing the extent of sorption.

Part I of this report showed the relationship between activity and particle size by fractionating soils dosed with zinc 65 and measuring the activity associated with each size fraction. The results indicated that the activity per gram on particles smaller than 43 microns in size was from 7 to 8 times greater than that on particles

from 43 to 250 microns. On this basis, it appeared desirable to determine whether a similar relationship existed for bottom sediments. If so, the inert bulk of sediment samples could be discarded and the small portion containing most of the activity could then be analyzed with greater ease and accuracy.

In addition to those described in Part I, various methods for size fractionation of soils and sediments have been described by Petelin *(2)*, Kilmer and Alexander *(3)*, and Sabo, Baptise, and Rueter *(4)*. These authors agree that materials larger than silt size (50 microns), as shown below, can most easily be separated into their respective size fractions by screening. Particles smaller than 50 microns may be sized by techniques employing elutriation, centrifugation, and sedimentation.

Classification of soil fractions *(4)*

Very fine sand	100 to 50 microns
Silt	50 to 2 microns
Clay	less than 2 microns

Kilmer *et al. (3)* point out that accurate size fractionation is dependent on complete dispersion so that no aggregates will be larger than the smallest size fraction to be separated. The methods of accomplishing this dispersion include: removal of flocculating ions, such as calcium; removal of cementing agents, such as organic matter and iron oxide; use of chemical dispersing agents; and mechanical agitation. It was shown in Part I that conventional dispersion methods employing chemicals should not be used on sediments before radioanalysis because the activity of the sample may be released by changes in physical-chemical equilibrium. Since any changes of these types would lead to erroneous results, two methods—mechanical vibrations by ultrasound and high-speed stirring—were investigated for physical dispersion of sediments.

These methods differ considerably from each other in the manner in which they achieve dispersion. Flammer *(5)* has reported that ultrasonic energy introduced into a suspension of sediments is dissipated by three mechanisms: development of shear waves at the liquid-solid interface because the particles vibrate in response to, but lag behind, the sound wave; reradiation by the particle causing a scattering of energy; and losses associated with heat conduction down thermal gradients. Hochman *(6)* noted that ultrasonic vibration produced extreme alternating accelerations and pressure differences of several thousand atmospheres over distances of one-half of a wavelength. The shear forces resulting from ultrasound would tend to cause aggregates of minerals and organic matter to tear apart.

In contrast, high-speed stirring causes dispersion of aggregates by attrition through the random grinding together of sediment particles. This abrasion should also tend to comminute flocculent organic matter and thereby release bound silt and clay particles.

EQUIPMENT AND METHODS

Ultrasonic dispersion was performed with the equipment shown in figure 1. The sonifier consisted of an ultrasonic generator and a probe-shaped converter or transducer, containing a cooling fan, active piezoelectric element, and a mechanical transformer. The converter was operated at a nominal frequency of approximately 20,000 cycles per second. The mechanical transformer was 1½ inches in diameter at the base and stepped down to one-half inch at the tip to form a probe about 2½ inches long.

Stirring was done with a variable speed electric motor stirrer equipped with a friction-cone drive adjustable from approximately 250 to 1,300 revolutions per minute. For this study, a 1½-inch diameter, four-blade propeller of stainless steel was used.

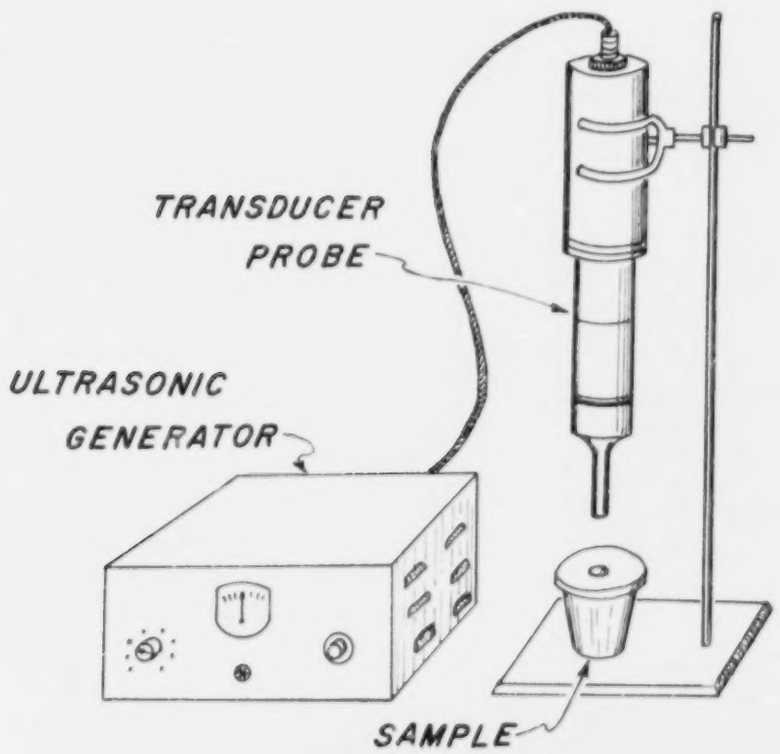


Figure 1. Ultrasonic dispersion equipment

The samples were dispersed in disposable 4½-ounce polypropylene containers with an inside diameter of 1¾ inches and a tight-fitting lid. Holes just large enough to insert the transducer probe or stirrer shaft were punched in the lids to prevent loss of the sample. The small diameter containers kept the sediments close to the probe or shaft for maximum agitation.

LABORATORY STUDY

Ultrasonic dispersion was evaluated in three phases during this study. First, the sediments were examined microscopically before and after dispersion. Second, a timed study was performed to determine the effect of sonification time on dispersion. Finally, sediments dosed with zinc 65 tracer were dispersed by ultrasound and stirring prior to size fractionation by screening. The main parameters for this evaluation were the oven-dry weight, the organic content, and the gamma activity of zinc 65.

MICROSCOPIC ANALYSIS

Sediment samples were examined at a magnification of 100 X before and after dispersion. Before dispersion there was a pronounced clumping of flocculent organic matter as shown in figure 2. These organic floccules entrapped many subangular particles ranging from about 5 to 50 microns in the largest dimension. Fewer than 5 percent of the particles in this size range were observed outside of the organic matrices.

Stirring for 10 minutes produced little observable change in physical appearance of the sediments shown in figure 2. The organic floccules were slightly smaller and more scattered but still prominent, with few of the trapped particles removed. Dispersion by ultrasound, however, caused significant visible changes. Figure 2 shows that no traces of the flocculent organic matter remained. All the clumps were dispersed and the silt and very fine sand-size particles were released so that each particle could be distinguished separately. The organic matter was subdivided into particles too small to be seen at 100 X and was distributed uniformly throughout the sample.

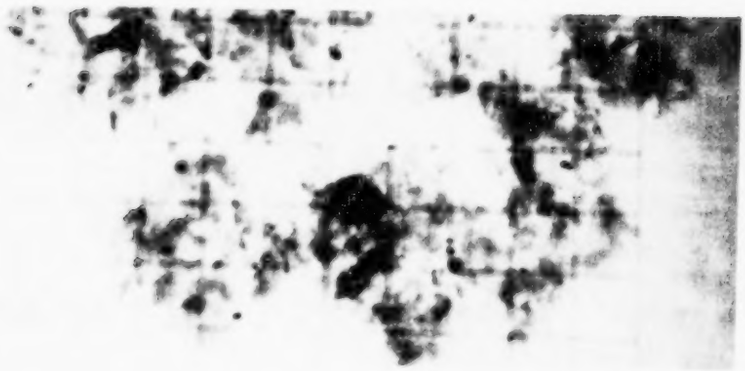
During dispersion treatment both methods appeared to impart a similar amount of agitation to the sediment particles, as viewed through the clear plastic container. After 10 minutes of treatment, however, the water with the sonified sediments had a dark brown muddy appearance, whereas the stirred samples were relatively clear.

EFFECT OF SONIFICATION TIME

Dispersion by ultrasound was further evaluated by observing the effects caused by duration of dispersion treatment on wet screening. Fifteen representative samples of river sediments, weighing 35

Photomicrograph of sediments before dispersion

Photomicrograph of sediments after stirring



Photomicrograph of sediments after ultrasonic dispersion

Figure 2. Microscopic analysis of sediments at a magnification of 100 X

grams each, were divided into five groups of triplicates to be dispersed for 0, 5, 10, 15, and 20 minutes, respectively. The three samples in each group were then wet screened on 250-, 125-, and 43-micron screens placed in series. Screening required rubbing with a rubber policeman for about 10 minutes while washing with about 400 milliliters of river water. Washing was continued until the water passing through the screens became clear. The fraction remaining on each screen, and the material smaller than 43 microns, removed from the wash water by centrifuging, were characterized by the oven-dry weight and the organic content. The organic matter was measured by the Walkley-Black chromic acid oxidation method *(7, 8)*. The centrifuged wash water was also analyzed for soluble organic matter by the COD procedure described in "Standard Methods" *(9)*.

The effect of sonification time on the weight of each size fraction is shown in figure 3. At zero sonification time (equivalent to stirred samples) more than 95 percent of the sediment particles remained on the 250-micron screen. Five minutes of sonification, however, resulted in a major change in distribution of each size fraction. The percentage of material greater than 250 microns dropped from about 95 percent to 90 percent of the total sample weight with a corresponding increase in the weight of the smaller size fractions. The fraction on the 125-micron screen increased about 2 percent after 5 minutes of dispersion and then remained nearly constant. The percentage of material on the 43-micron screen increased with longer sonification time as did the material smaller than 43 microns. Smaller changes in weight were noted in each case after 10 to 15 minutes of treatment.

The organic content of each size fraction also varied considerably with the time of sonification as indicated in figure 4. Before dispersion, 33 percent of the organic matter was associated with the 250-micron fraction, and 48 percent of the organic matter was associated with the fraction less than 43 microns. Again, the most significant change occurred within the first 5 minutes. The organic matter on the 250-micron screen dropped about 24 percent after 5 minutes of treatment. In the same time, the organic content of the wash water, which was initially about 6 percent, increased to 23 percent. After 15 minutes of dispersion, about 58 percent of the total organic matter was associated with the fraction less than 43 microns and about 34 percent remained in the centrifuged wash water. Together, these two fractions contained 92 percent of the total organic matter. An additional 5 minutes of treatment produced no noticeable changes in the distribution of the organic matter.

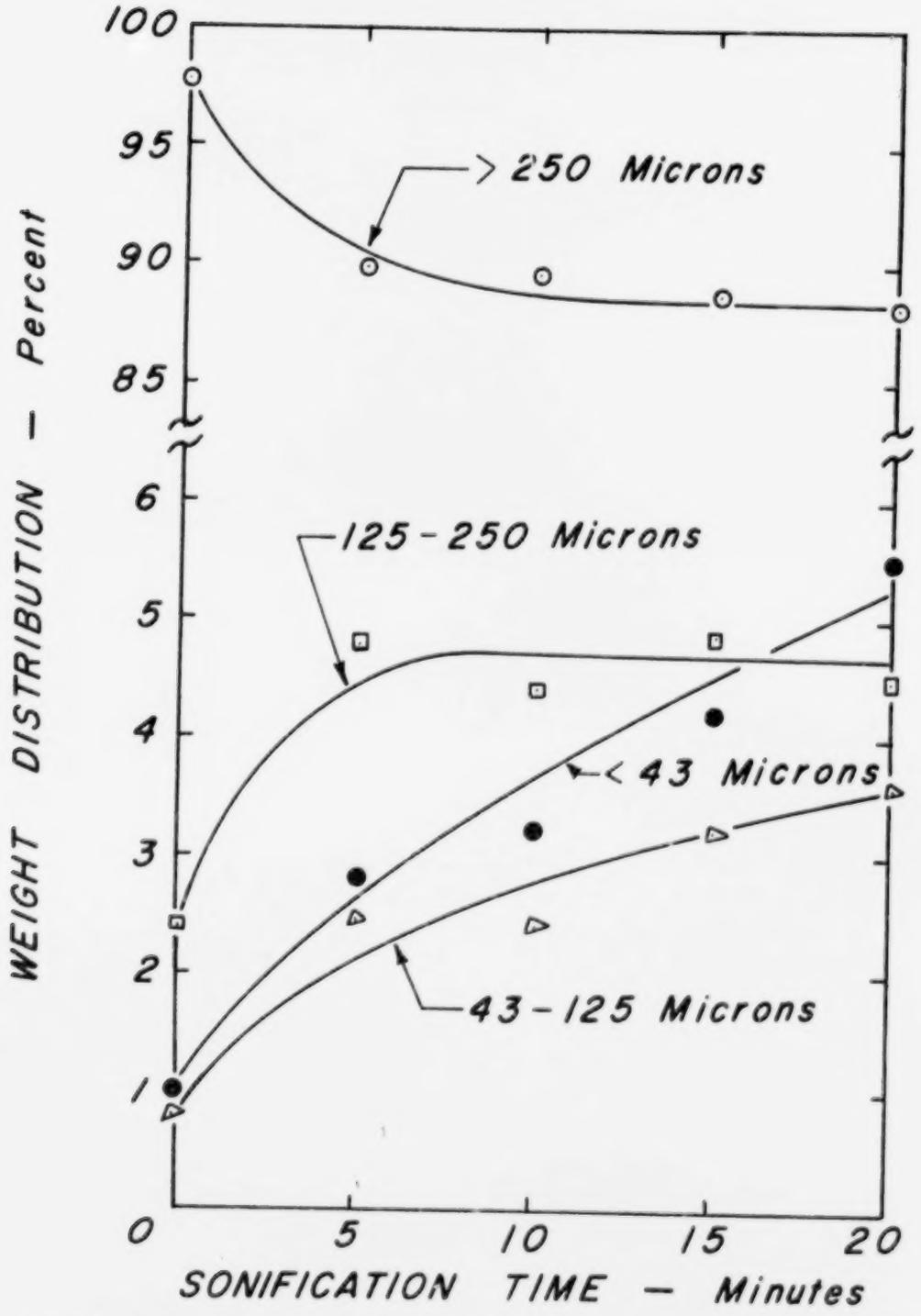


Figure 3. The effect of sonification time on weight distribution of sediment fractions

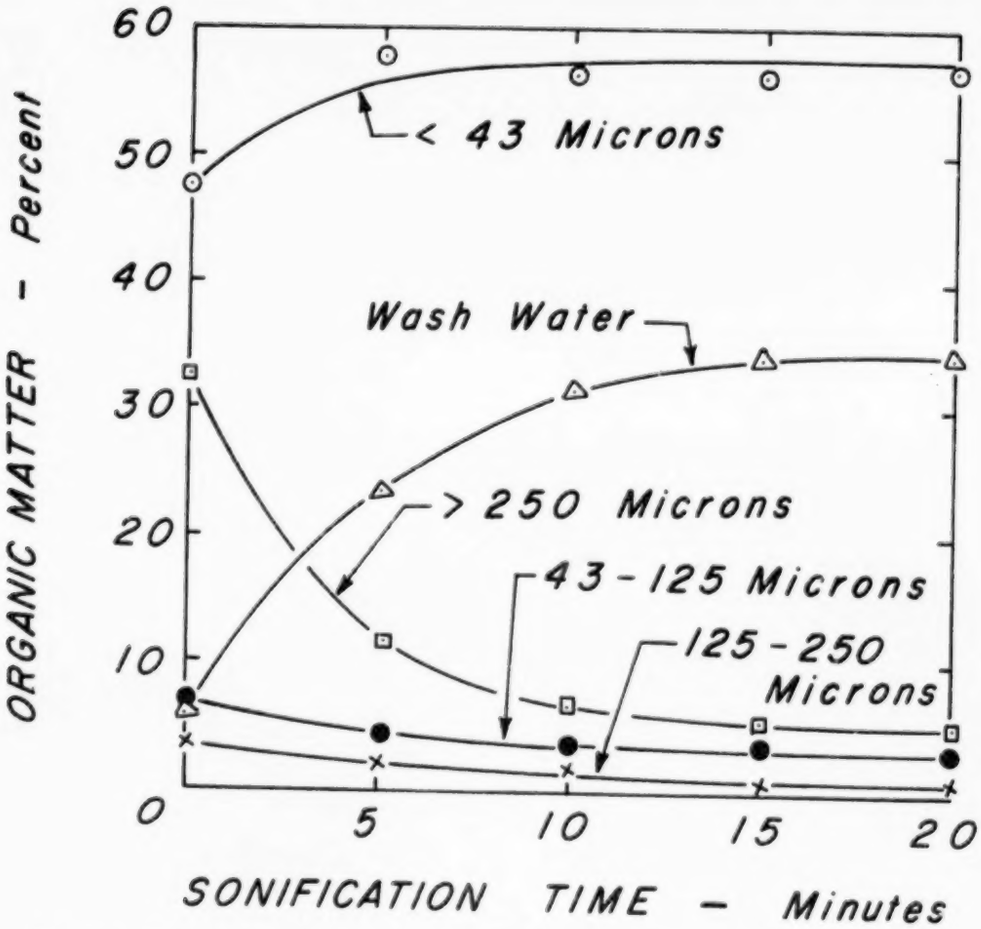


Figure 4. The effect of sonification time on the organic content of sediment fractions

The results of both the weight and organic content analyses show that the degree of dispersion was highly dependent on the duration of treatment. Even 5 minutes of dispersion with ultrasound allowed a much better separation of size fractions than stirring. In this study, samples weighing about 35 grams required 15 to 20 minutes of ultrasonic dispersion for optimum size fractionation. In general, the optimum sample size and treatment time would depend on the power output capacity of any given ultrasonic generator.

RADIOANALYSIS OF SPIKED SEDIMENT FRACTIONS

The material used in this part of the study consisted of bottom sediments and water collected from the Concord River, near Concord, Massachusetts. About 4.5 kilograms of sediments, consisting of particles smaller than 3,300 microns, were placed in each of four glass aquaria with 4 liters of river water. These samples were dosed with 0.025, 0.05, 0.2, and 0.6 microcuries of zinc 65, respectively. The contents of each aquarium were mixed and allowed to

settle for 3 days. Then 10 samples ranging from about 5 to 60 grams were scooped randomly from each aquarium. Each sample was analyzed by gamma-scintillation spectrometry for the amount of zinc 65 activity present. After counting, the 10 samples from each aquarium were divided into two groups of five according to activity so that each group would have a similar range. One group was then dispersed with ultrasound and the other by mechanical stirring, prior to size fractionation.

After dispersion, the sediments were rubbed on a Tyler standard screen with openings of 43 microns while washing with river water in the same manner as with the previous timed duration study. In this way, the sediments were separated into a coarse fraction of particles larger than 43 microns and a fine fraction of particles smaller than 43 microns. The fine fraction was removed from the wash water by centrifuging for 10 minutes at about 2,100 g.

Both size fractions were analyzed for zinc 65 to determine the proportion of the original activity associated with each. The centrifuged wash water was also analyzed for zinc 65 to indicate whether any activity was released from the sediments by the dispersing treatment. The weight of the size fractions was determined after drying overnight at 110° C.

Results obtained from this phase of the study are summarized in table 1. The weights of the two groups of bulk samples were about 24 and 27 grams. After dispersion and screening, sonification allowed about 6 percent of the bulk sample to be separated as fines, whereas stirring released only 2 percent or only one-third as much. This indicates that ultrasonic dispersion permitted better separation for accurate size fractionation.

Table 1. Summary of activity and dry weight data

Dispersion method	Bulk weight grams	Coarse fraction (43–3,300 microns)		Fine fraction (< 43 microns)		Wash water
		Weight percent	Activity percent	Weight percent	Activity percent	Activity percent
Ultrasound	23.93	94	23	6	54	23
Stirring	27.32	98	62	2	32	8

The activity analyses indicate that after ultrasonic dispersion, 54 percent of the activity was associated with the fine fraction. This was especially significant since this fine fraction comprised only 6

percent of the sample. By comparison, after stirring, most of the activity (62%) remained with the coarse fraction. This was nearly three times the activity that remained with the coarse fraction dispersed by ultrasound.

The activity per gram of fine fraction was about 75 times the activity per gram of coarse fraction after ultrasonic dispersion. This shows that small changes in the weight of the fine fraction could be responsible for large variations in the activity of unseparated bulk samples. Also, this portion may be analyzed separately and used as a basis for comparison of sediments widely different in composition. Furthermore, the high activity and small volume of the fine fraction allow more efficient counting for greater accuracy.

The 23 percent activity in the wash water (table 1) after ultrasonic dispersion was not released from the sediment particles. This activity was mainly associated with organic matter subdivided by dispersion into particles too small to be removed from the wash water by centrifugation.

A comparison of the results after dispersion by ultrasound and by stirring shows that radioanalyses of size fractions may vary widely depending on the method of treatment and degree of dispersion. In this study, ultrasonic dispersion produced better size fractionation, permitting meaningful radioanalysis of the fractions.

Ease of screening was also enhanced by ultrasonic dispersion. Screening was performed more quickly with less washing and rubbing. It was also easier to determine when fractionation was complete.

FIELD STUDIES

The ultrasonic dispersion technique has been applied on sediment samples collected above and below several nuclear facilities discharging radioactive wastes. These sediments were first dispersed by ultrasound and then separated into two fractions with a 43-micron screen. Radioanalysis was performed only on the fraction less than 43 microns because this fraction contained essentially all the activity in each case. Using this technique the following radionuclides were identified by gamma spectrometry: ^{144}Ce, ^{232}Th, ^{226}Ra, ^{51}Cr, ^{125}Sb, ^{106}Ru, ^{137}Cs, ^{134}Cs, ^{54}Mn, ^{65}Zn, ^{60}Co, and ^{40}K. Identification of small quantities of some of these radionuclides was aided by the reduced sample volume and improved counting geometry achieved by discarding the relatively inert fraction of the seiments greater than 43 microns.

The major advantage of this approach is the reduction in variability of sediment properties made possible by considering only the fraction less than 43 microns. In this way the effect on radionuclide content due to sediment composition was minimized and comparisons could be made between sediments collected at different times and places. Ultrasonic dispersion makes radioanalysis results more reproducible by assuring uniform separations of sediment fractions from samples of widely varying composition.

SUMMARY AND CONCLUSIONS

The physical changes imposed on the sediment samples by treatment with ultrasound and mechanical stirring were evident from the microscopic analyses. Stirring caused little or no dispersion of the binding matrix. Ultrasound, on the other hand, appeared to completely disperse the flocs with the release of all the bound silt and clay particles. Further evidence of physical change was the ease of screening after dispersion. The samples treated by ultrasound required little rubbing or wash water to obtain a clear effluent from the screen. This was possible because most of the particles smaller than 43 microns were released from the binding matrix. In contrast, stirring did not break up the organic flocs and considerable rubbing and washing was needed to obtain a clear effluent.

The second phase of this study indicated that the duration of ultrasonic treatment had a significant effect on the degree of dispersion. Both the weight and the organic content of the larger size fractions decreased with sonification time. Thus, as dispersion time increased, more particles were separated from the organic matrix and passed through the screens. In addition, the organic matrix was dispersed into colloidal sizes that were not removed from the wash water. These parameters did not change significantly beyond a sonification time of 15 to 20 minutes for sediment samples of about 35 grams. Longer treatment time would be required, however, for samples larger than 35 grams or for sonifiers of lower power output.

The tracer study with zinc 65 showed that particles smaller than 43 microns had 75 times more activity per gram than the coarse fraction. Therefore, poor size separation might yield large errors in activity measurements for sediment fractions. For example, stirring did not allow good size separation and thus most of the activity remained with the coarse fraction. By comparison, dispersion by ultrasound would assure better separation of the sedi-

ment particles into the required size fractions. Then radioanalysis of these fractions would give meaningful results with confidence that each fraction contained only particles of the size range specified.

The main conclusions of this study were:

1. Ultrasonic dispersion allowed fractionation of sediments into desired size ranges for radioanalysis by releasing particulates from binding organic floccules.
2. Stirring did not disperse sediments adequately for separation into accurate size fractions.
3. Ultrasonic dispersion made the screening process faster (by requiring less rubbing and washing); moreover, it was easier to determine when fractionation was complete.
4. Sediment samples of about 35 grams required 15 minutes of treatment with ultrasound for good dispersion.
5. Ultrasonic dispersion and separation of the fraction smaller than 43 microns made sediment analyses reproducible so that samples of widely varying compositions could be compared.

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